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APPENDIX E

NTRD DOCUMENT COVER SHEET [1]

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This deliverable was prepared in accordance with Los Alamos National Laboratory
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QA program which meets the requirements of
☒ DOE Order 414.1 ☐ NQA-1 ☐ Other

This Deliverable was subjected to:

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Assessment of thermophysical property characterization of MiniFuel scale geometries

Nuclear Technology Research and Development

Prepared for
U.S. Department of Energy
Advanced Fuels Campaign
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Los Alamos National Laboratory

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SUMMARY

In this report, thermophysical characterization of a set of test specimens adopting MiniFuel geometries is described. Specific heat capacity and thermal diffusivity of metallic samples (copper, 304 stainless steel, and Inconel 600) and two nuclear fuel samples (UO_2 and U_3Si_2) are measured. The primary aim is to validate and assess the feasibility of thermophysical characterization of fuels with MiniFuel geometries that will be irradiated at the High Flux Isotope Reactor (HFIR) at Oak Ridge National Laboratory (ORNL).

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ACRONYMS

AFC	Advanced Fuels Campaign
ATF	Accident tolerant fuel
LANL	Los Alamos National Laboratory
HFIR	High Flux Isotope Reactor
ORNL	Oak Ridge National Laboratory
DSC	Differential scanning calorimetry
LFA	Laser flash analysis

1. INTRODUCTION

Ongoing efforts within the Advanced Fuels Campaign (AFC) have involved synthesis and characterization of accident tolerant fuels (ATF). Uranium silicide (U_3Si_2) has been proposed as a candidate material as it offers enhanced thermal conductivity (this reduces the propensity for thermal stress failures) and a uranium density that is higher than conventional uranium dioxide fuels, which can enhance the economic viability of nuclear power plant operation. Nevertheless, full elucidation of the thermophysical properties of U_3Si_2 at peak operating temperatures is necessary for any potential implementation in the current nuclear energy infrastructure.

For nuclear energy applications, development of U_3Si_2 thermophysical properties databases must include pre and post-irradiation scenarios. Prior efforts, conducted at Los Alamos National Laboratory (LANL), supported by the AFC have yielded detailed assessments of unirradiated U_3Si_2 thermophysical properties at temperatures up to 1500 °C [1]. However, conducting similar measurements on irradiated fuel specimens poses numerous challenges. Primarily, irradiated samples are highly radioactive and can make handling prohibitively expensive. Additionally, full scale integral effects testing of fuel yields multiple coupled intrinsic properties that are difficult to deconvolute. To address these problems, experimental designs (Figure 1) have been developed and realized at Oak Ridge National Laboratory (ORNL) to conduct irradiation experiments on miniature fuel (MiniFuel) specimens at the High Flux Isotope Reactor (HFIR) [2]. The proposed dimensions for the MiniFuel design are diameters ~3 mm and thickness less than or equal to 300 μm.

A test plan is currently being carried out at LANL to fabricate fresh nuclear fuel specimens with MiniFuel scale geometries [3]. Concurrently, the viability of characterizing thermophysical properties will be evaluated, given the size restraints imposed by miniature scale geometries. Recently, thermal diffusivity has been validated on miniature W specimens using a “Hyper Flash” thermal analyzer [4]. In this report, the heat capacity and thermal diffusivity (using more conventional techniques) on standard metallic samples (copper, 304 stainless steel, and Inconel 600) adopting MiniFuel geometries are also described. The metals were chosen to provide a range of thermal diffusivity values to assess feasibility of the LFA system at LANL. Preliminary thermophysical characterization on a MiniFuel UO_2 and U_3Si_2 specimens are also discussed.

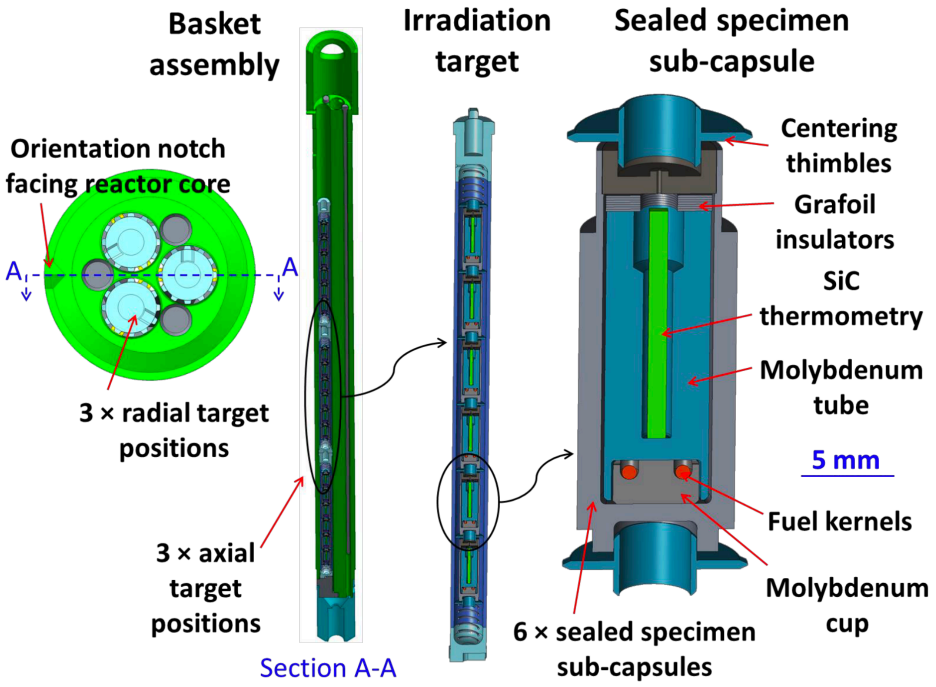

Figure 1: MiniFuel irradiation test setup developed at ORNL [2].

2. Experimental Setup

2.1 Specimen Synthesis and Fabrication

MiniFuel copper, 304 stainless steel, and Inconel 600 pellets (3 mm diameters and ~300 µm thickness) were fabricated from stock material using a wire electrical discharge machine (EDM)—see Figure 2 a-c. For UO_2, a previously synthesized 3 mm pellet with ~400 µm thickness was used for initial testing of a nuclear fuel specimen. A MiniFuel U_3Si_2 pellet (Figure 2 d) was made via powder (~0.25 wt% of ethylene-bisstearamide was added as binder) originating from an arc-melted button, where it was processed in an Ar glovebox line with oxygen content was maintained below 30 ppm. The powder was sieved to achieve a particle size less than or equal to 45 µm and ~0.10-0.15 g was loaded to a 3 mm punch and die set and pressed to ~300 MPa to form a pellet. The green pellet was loaded in W metal furnace enclosed within the glovebox line and sintered at 1460-1470 °C for 12 h in an Ar atmosphere. Geometric measurements on the as sintered pellet showed a diameter of ~2.8 mm and a thickness of ~1 mm. The pellet was then ground to ~255 µm thickness using 400-grit SiC paper. Masses and thicknesses of all samples are summarized in Table 1.

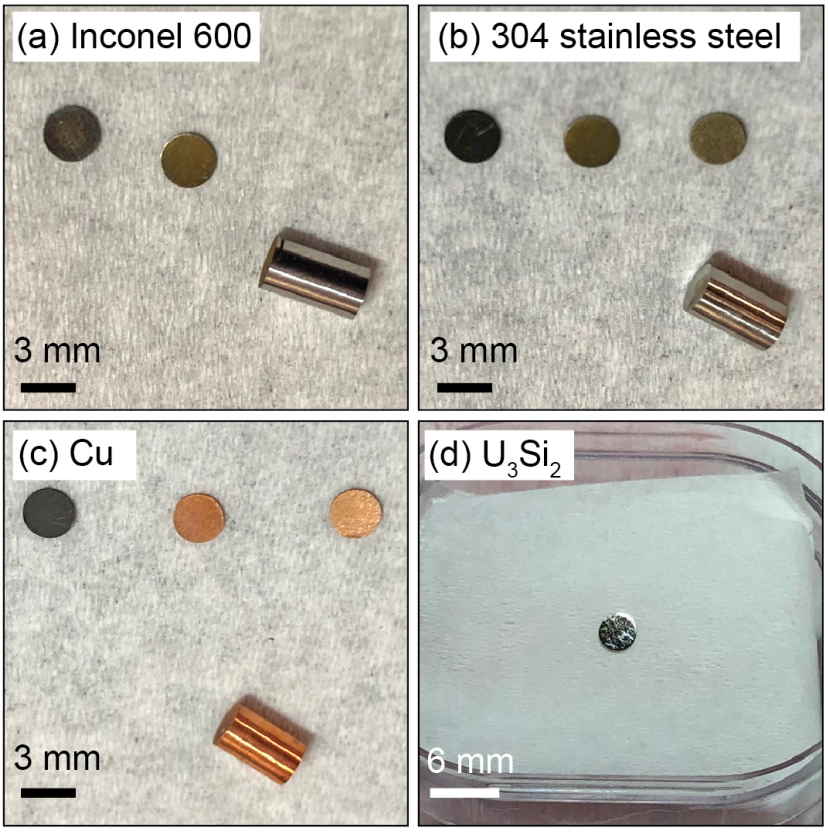

Figure 2: (a-c) Metallic samples cut into MiniFuel geometries using a wire EDM. (d) U_3Si_2 MiniFuel samples processed at LANL. The black surfaces on the Cu and stainless steel specimens originate from graphite coating need for LFA measurements.

Table 1: Summary of measured masses and thickness of fabricated pellets.

Specimen	Mass [mg]	Thickness [μm]; ± 1 μm
Cu	18.10	294
304 stainless steel	16.79	304
Inconel 600	16.34	316
U_3Si_2	18.95	255
UO_2	25.59	400

2.2 Differential Scanning Calorimetry (DSC)

Specific heat capacities of MiniFuel specimens were determined via the ratio method using differential scanning calorimetry adhering to the ASTM E1269 standards. (DSC; Pegasus 404C, Netzsch Instruments, Germany). Samples in the calorimeter were contained in Pt pans lined with Al_2O_3 to prevent pan-specimen reactions at higher temperatures (temperature was raised to 700 °C at a rate of 20 °C/min). An Ar atmosphere was maintained during the DSC measurements, where it was flowed through a Cu getter to reduce oxygen concentration in the sample chamber to ~10^{-1} to 1 ppm O.

2.3 Laser Flash Analysis (LFA)

Room-temperature thermal diffusivity was measured using laser flash analysis (LFA; LFA 427, Netzsch Instruments, Germany). Specimens were loaded into the LFA using a machined graphite fixture designed to accommodate MiniFuel geometries (Figure 2). A pulsed laser (0.30 ms pulse width) heats the lower surface of the sample and temperature change on the upper surface is then measured by an infrared detector. Thermal diffusivity can then be inferred from sample thickness and the time measured to reach half-maximum temperature change ($t_{1/2}$) by fitting the temperature rise signal with a modified Cowan model [5, 6]. The upper and lower surfaces of the samples were coated with graphite (Graphit 33, Kontakt Chemie, Germany) to improve emissivity and energy absorption from the laser pulse, respectively (the added graphite had minimal impact on thickness as no appreciable mass change in coated pellets was detected). Samples were pulsed with the laser 10 times for improved statistics.

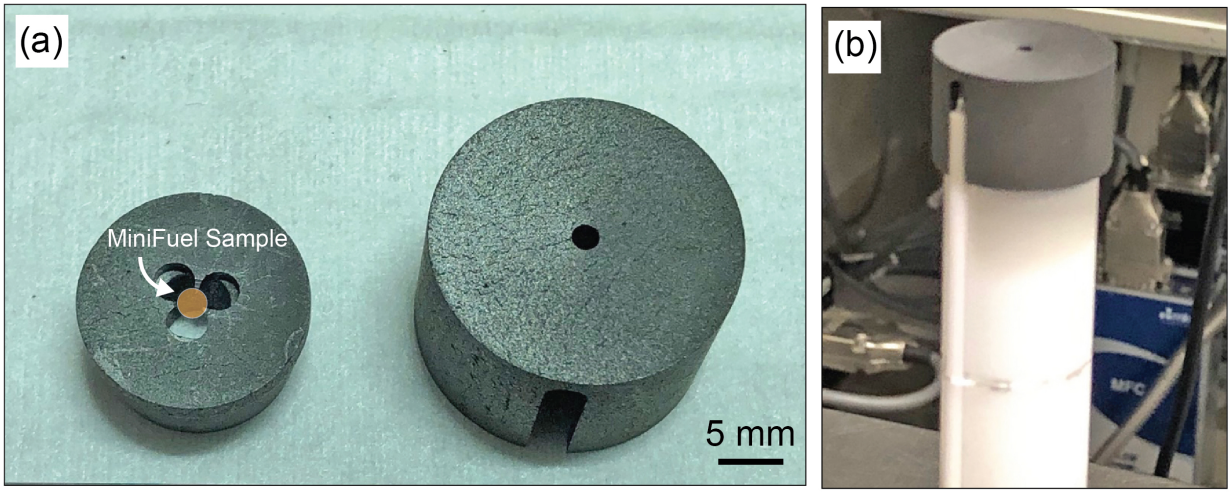

Figure 3: (a) Fabricated LFA fixture designed to accommodate MiniFuel specimens. (b) Fixture-specimen assembly mounted on the LFA instrument.

3. Results and Discussion

3.1 Specific Heat Capacity

Reference and measured specific heat capacities up to 700 °C of Cu, Inconel 600, and U_3Si_2 are shown in Figure 4. Overall, heat capacities stayed relatively constant with increasing temperature (one must note, however, the narrow temperature range). In comparison with the literature (obtained from a standards library in the DSC instrument for the metallic samples and the HSC database for U_3Si_2), data for U_3Si_2 and Inconel 600 were below the reference trends. Conversely, the measured Cu specific heat capacities were consistently higher than the reference. The discrepancies illustrates the difficulty in extracting heat capacity values from low mass miniature samples, where DSC signal to noise ratio is relatively low and thus can reduce the precision of heat capacity calculations. This problem is magnified for heavier actinide samples, which generally have low specific heat capacities. Nevertheless, there are feasible engineering solutions that will be explored that include using smaller specimen crucibles that may aid in reducing background signals.

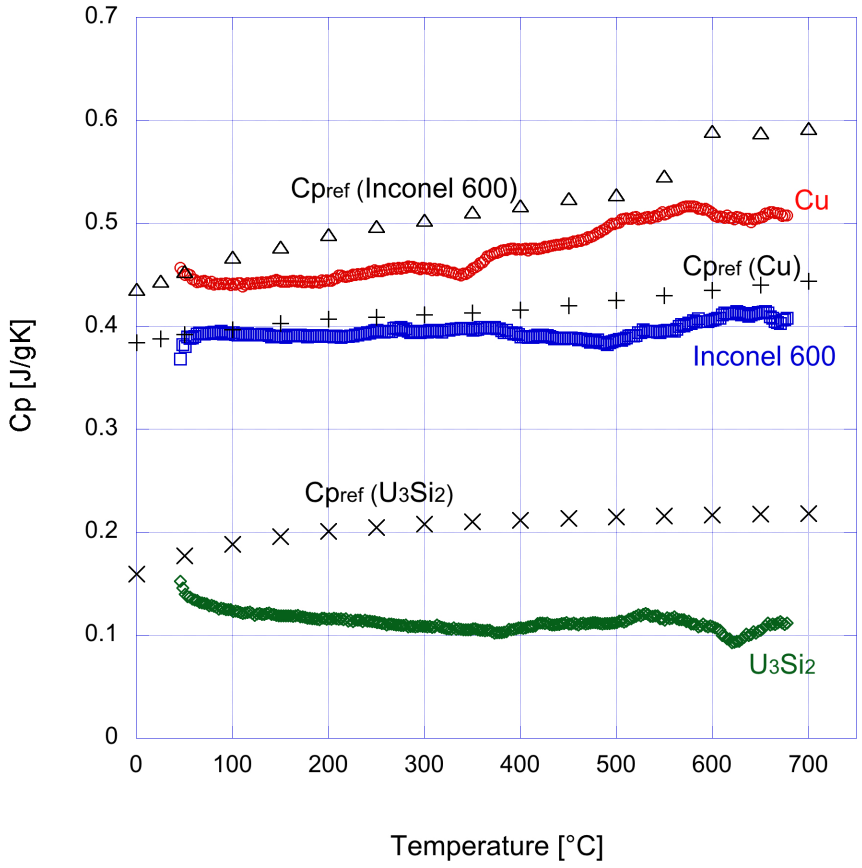

Figure 4: Measured and reference specific heat capacities of Cu, Inconel 600, and U_3Si_2 plotted against temperature.

3.2 Room-Temperature Thermal Diffusivity

Room-temperature thermal diffusivity data for all specimens tested are listed in Table 1. A large discrepancy is observed in Cu, which is known to have a high thermal conductivity. Here, measured values are an order of magnitude lower than the reference thermal diffusivity. Nevertheless, differences between measured and reference room-temperature thermal diffusivities become smaller as the thermal conductivity of the specimen decreases. It is important to note, however, that thermal conductivities will change with temperature variations, where the behavior is dictated by the nature of the material. Interestingly, measured thermal diffusivity of U_3S_2 at room temperature closely matches previously measured values [1].

The inconsistencies observed in the thermal diffusivities are tied to the underlying principles of the laser flash method. Infrared signals emitted after sample heating are relatively low at room temperature, which can give rise to large variations in measured thermal diffusivities (infrared signal generally improves with increasing temperature). Additionally, an accurate measurement requires a laser pulse width that is faster than the diffusion time of heat in a given specimen. It follows that very thin samples will quickly diffuse heat and thus require a very narrow laser pulse. Generally, the width of the laser pulse must be less than 1/10 of the time to reach half-maximum temperature ($t_{1/2}$) change in the specimen (guideline based on ASTME E1461). The LFA 427 instrument used in this study has a minimum laser pulse of 0.3 ms. As a result, the minimum $t_{1/2}$ value that can theoretically be measured is 3 ms. This is illustrated in Figure 5, which shows the temperature rise (inferred by the infrared detector signal) for the U_3Si_2 MiniFuel specimen. The $t_{1/2}$ value that is extracted from the Cowan model is ~3.6 ms (the model shows reason agreement with the measured signal, with a computed R^2 value of ~0.99312). This approaches the instrumental limitation of the LFA427 system that is available at LANL. Accordingly, the minimum thermal diffusivity (α_{min}) than can measured for a sample with a given thickness (L) can be deduced from the Parker equation [7]:

$$\alpha_{min} = 0.1388 \left(\frac{L^2}{t_{1/2}} \right)$$

For a sample with a 300 μm thickness, the minimum thermal diffusivity that can be measured in the LFA 427 instrument is ~4.16 mm^2/s. Hence, measurements of nuclear fuel materials such as U_3Si_2 are beyond the resolution capable of the instrument that is currently being deployed. Hence, accurate measurements of thermal diffusivity on MiniFuel specimens will require laser pulses that are at least an order of magnitude less than what is currently available in the LFA 427 instrument.

Table 2: Measured room-temperature thermal diffusivities measured via LFA compared with reference values.

Specimen	α(measured) [mm²/s]	α(reference) [mm²/s]
Cu	8.733 ± 0.442	111 [9]
304 stainless steel	5.193 ± 0.106	4.2 [10]
Inconel 600	5.647 ± 0.255	3.428 [11]
UO_2	8.080 ± 0.134	3.3 [12]
U_3Si_2	3.602 ± 0.056	3.6 [1]

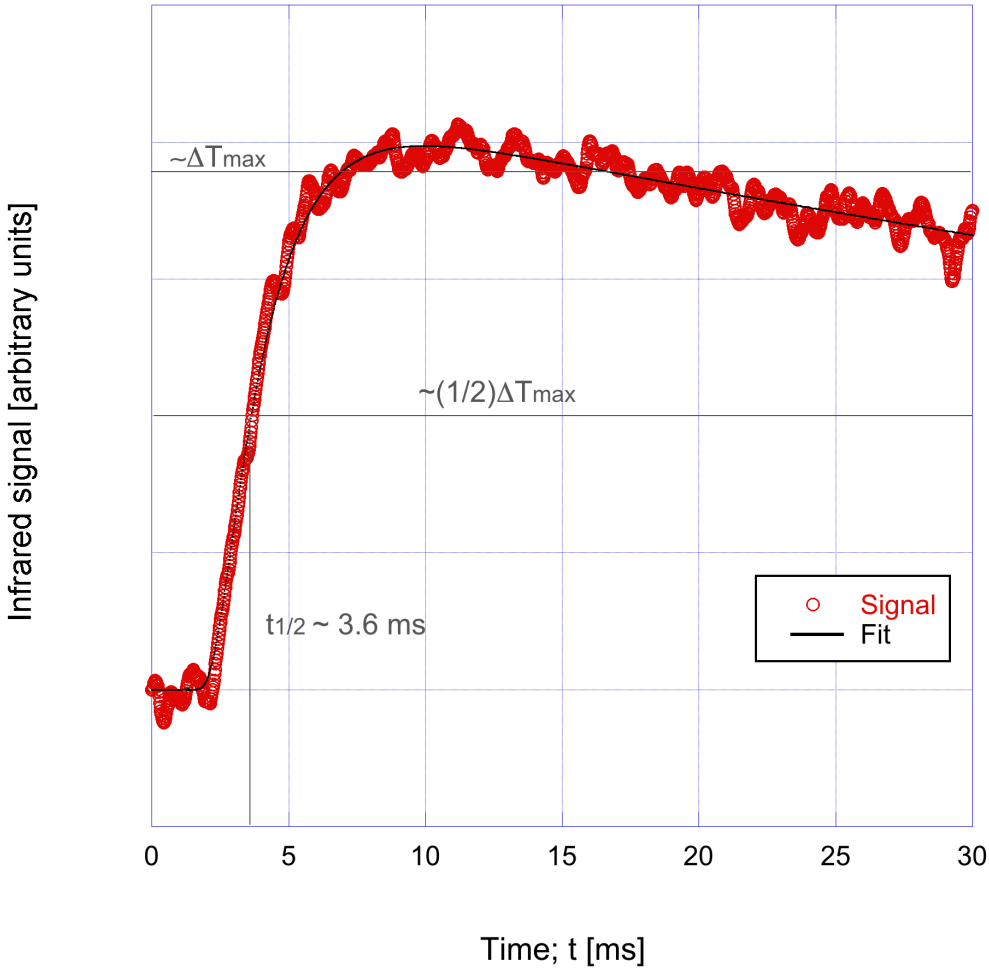

Figure 5: Infrared detector signal representative of the rise of a U_3Si_2 MiniFuel sample temperature after being exposed to a laser pulse. The time to reach half the maximum temperature change is ~3.6 ms (based on the Cowan model), which approaches the instrumental limit (~3 ms) that is capable of being measured on the LFA 427.

4. Conclusions and Next Steps

In this report, preliminary results on thermophysical property measurements on miniature test specimens are presented. Materials such as Cu, 304 stainless steel, Inconel 600, and U_3Si_2 have been successfully fabricated to MiniFuel geometries. Specific heat capacities measured via differential scanning calorimetry (DSC) give deviate from the reported values in the scientific literature. Given the limitations of deducing heat capacities of low-mass samples, the initial results are expected and may be improved upon by modifying the current DSC setup to accommodate miniature specimens. Thermal diffusivities measurements of MinFuel specimens prove to be more difficult given the instrument limitation of the laser flash analysis (LFA) setup that is currently being used at LANL. Nevertheless, the issue can be addressed by employing a laser that is capable of pulses faster than the diffusion of heat in a thin MiniFuel sample. Such lasers are currently provided by manufacturers of LFA instruments.

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